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The uncertainties of satellite DOAS total ozone retrieval for a cloudy sky

A.A. Kokhanovsky *, V.V. Rozanov

Institute of Environmental Physics, University of Bremen, Otto Hahn Allee 1, D-28334, Bremen, Germany

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Abstract

This paper aimed to the study of the influence of the uncertainties of satellite Differential Optical Absorption Spectroscopy (DOAS) total ozone retrieval scheme due to unconstrained parameters of the plane-parallel scattering cloud model. The work is based on the exact radiative transfer calculations for a realistic atmospheric model described in the paper. In particular, the accuracy of the DOAS technique based on hyperspectral measurements in the ozone absorption band 315–340 nm is investigated. The general weighting functions approach to the calculation of the error of total ozone introduced in the paper can be used for studies of errors in the retrieved concentration of other trace gases (e.g., NO_2, SO_2, etc.) as well. It was found that the uncertainties in the cloud geometrical and optical characteristics can produce absolute errors in the retrieved total ozone column of up to 6%. This calls for the introduction of accurate cloud retrieval algorithms in ozone satellite retrieval schemes.

Keywords: Radiative transfer; Light scattering; Ozone retrievals; Clouds

1. Introduction

The monitoring of variations in concentrations of the atmospheric ozone is of great importance for environmental studies. The range of variability is 220–490 DU with ozone maximum located somewhere in the range 16–25 km (Lamsal et al., 2004). Usually, ozone concentration measurements are performed from ground using Dobson spectrophotometers (DS) or Brewer spectrometers. The accuracy of ground-based measurements using DSs is about ±2%.

UV total ozone retrievals similar to the ground-based Dobson technique have been extended to satellite measurements of backscattered solar light. The accuracy of satellite measurements is lower as compared to the ground-based techniques due to a number of a priori assumptions in the correspondent retrieval algorithms. However, they have much better spatial coverage and quite high precision. In particular, Weber et al. (2005) found that RMS between ground and satellite measurements is typically just ±5%. Therefore, satellites have a great and well demonstrated potential in the derivation of total ozone concentrations on a global scale. This is especially important for remote areas like Arctic and Antarctic.

Series of TOMS (Total Ozone Mapping Spectrometer) (Herman et al., 1997; Niu et al., 1992; Ahmad et al., 2004), SAGE (Stratospheric Aerosol and Gas Experiment) (McDermid et al., 1990), and SBUV (Solar Backscattered Ultraviolet Stratospheric ozone) (Mateer

* Corresponding author. Tel.: +49 421 218 2915; fax: +49 421 218 4555.
E-mail address: alexk@iup.physik.uni-bremen.de
(A.A. Kokhanovsky).

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et al., 1971; Klenk et al., 1982; McPeters et al., 1994) provided a wealth of useful information on total ozone distributions around the globe. These measurements were complemented by retrievals from Global Ozone Monitoring Experiment (GOME) (Burrows et al., 1999; Koelemeijer and Stammes, 1999; Weber et al., 2005), Scanning Imaging Absorption Spectrometer for Atmospheric CHartographY (SCIAMACHY) (Bovensmann et al., 1999) and Ozone Monitoring Instrument (OMI) (Levelt et al., 2005). The algorithms to retrieve the total ozone from GOME and SCIAMACHY are based on the analysis of differential structure of backscattered signal in the Huggins bands. Such techniques are called DOAS (Differential Optical Absorption Spectroscopy). The slowly varying background component, which is largely due to atmospheric scattering and surface reflection effects, is subtracted from the registered signal using correspondent polynomial fitting techniques. This gives a major difference with earlier developed techniques based on the analysis of ratios of absolute intensities of solar backscattered light at several wavelengths.

The influence of clouds on ozone retrievals using, e.g., TOMS has been studied by Newchurch et al. (2001), Liu (2002), Liu et al. (2004) and Ahmad et al. (2004) among others. Koelemeijer and Stammes (1999) performed correspondent investigations for GOME UV hyperspectral measurements.

It is generally understood now that clouds have

- albedo effect (AE),
- increase in-cloud absorption effect (CAE),
- shielding effect (SE) on ozone retrievals.

The first two effects (AE and CAE) lead to the apparent increase of the depth of absorption bands of ozone (and, therefore, the overestimation of total ozone). The third effect leads to the underestimation of the vertical ozone densities.

Indeed, clouds increase the amount of reflected light due to the generally larger cloud albedo as compared to the clear sky case. So the retrieved total ozone is overestimated due to more absorption of light by ozone above clouds. This is related to the fact that more light is available for absorption by ozone then (not only direct light but also light backscattered from a cloud is of importance). Of course, such an increase in absorption is a solely cloud presence effect. This is in no way related to the enhanced ozone production (e.g., in stratosphere).

Increase in-cloud absorption effect is due to the fact that radiation travels between droplets in clouds several times leading to the increase of absorption by ozone on the way between droplets (as compared for the same path in a cloudless atmosphere). This effect is purely due to light scattering and also not due to the ozone enhanced production in a cloud. If in-cloud scattering is not accounted for in a correct way in the retrieval algorithm, then the overestimation of total ozone by a retrieval scheme takes place.

The last effect (SE) is due to the fact that less photons penetrate underneath the clouds leading to the "apparent" decrease of depths of absorption lines. This is not due to the "ozone hole" but solely due to the fact that less light is available for absorption.

Although the physics of influences of clouds on the total ozone error is well understood, the quantitative estimates of errors depend on a particular algorithm used in the retrieval.

The aim of this paper is to study the influence of uncertainties in cloud properties (e.g., cloud top height, geometrical and optical thicknesses) on the total ozone column retrievals using space-borne hyperspectral measurements in the framework of DOAS technique. Here, only effects of horizontally homogeneous cloud fields on the total ozone retrievals are studied. We emphasize the importance of simultaneous accurate cloud and ozone retrievals using spectral measurements of the TOA reflectance (e.g., in the UV and in the oxygen A-band) for a given atmospheric state. Then the retrieved cloud information can be used for the improved determination of total ozone.

2. Total ozone retrieval error and its relationship with weighting function

The estimation of errors in the retrieved total ozone column due to the uncertainties in the cloud properties can be performed in the following way. At first the forward radiative transfer model runs for a given ozone concentration. This enables the determination of the backscattered spectral reflectance for a given modeled cloud system (e.g., with a fixed cloud top height). Then this spectrum is used in the retrieval procedure, where the same cloud properties as in the forward model are assumed. This run is needed to ensure that the retrieved ozone concentration coincides with the total ozone assumed in the forward model. The next step is perturbation of a given cloud parameter (e.g., cloud top height) and a new run of the retrieval. The difference between two retrievals gives the theoretical error of uncertainty in a cloud parameter on the total ozone error. Such a technique requires multiple runs of retrieval algorithms.

It is possible to derive the correspondent errors in yet another way using the fact that the studied error in the total ozone can be related to the correspondent

weighting function (WF). In this section we derive the correspondent relationship between the error and WF. The derived equation is used in all numerical experiments performed in this work. The nature of absorbing component is not specified in the theoretical derivations. So the results obtained in this section are very general and can be used for any trace gas.

To start with, we note that the reflectance R at the wavelength λ depends nonlinearly on the ozone vertical profile. To retrieve the total ozone, the following approximate linear relationship between the reflectance spectrum corresponding to a priori assumed vertical ozone profile $n_0(z)$, denoted $R\left(\lambda,n_0(z),\overrightarrow{p_t}\right)$, and the spectrum $R\left(\lambda,n_t(z),\overrightarrow{p_t}\right)$, corresponding to the perturbed profile

$$n_t(z) = n_0(z) + \delta n(z), \tag{1}$$

is used:

$$R\left(\lambda,n_t(z),\overrightarrow{p_t}\right) = R\left(\lambda,n_0(z),\overrightarrow{p_t}\right) + \int_0^H W\left(\lambda,n_0(z),\overrightarrow{p_t}\right)\delta n(z)dz. \tag{2}$$

Here $R\left(\lambda,n_0(z),\overrightarrow{p_t}\right)$ is the simulated reflectance spectrum (the synthetic spectrum) corresponding to the atmospheric parameters $\left\{n_0(z),\overrightarrow{p_t}\right\}$, H is the TOA altitude (equal to 60 km in this study), $W\left(\lambda,n_0(z),\overrightarrow{p_t}\right)$ is the so-called weighting function (Rozanov et al., 1998) providing the desired linear relationship between the measured spectra and the variation of the vertical ozone profile, i.e., $\delta n(z)$. We assume in this work that the vector parameter $\overrightarrow{p_t}$ has three components. They are cloud top height h_{top}, cloud bottom height h_{bot}, and cloud optical thickness τ. Actually, the exact number and actual meaning of components is of no importance for the theory described below.

The functions $R\left(\lambda,n_0(z),\overrightarrow{p_t}\right)$ and $W\left(\lambda,n_0(z),\overrightarrow{p_t}\right)$ in Eq. (2) can be calculated using the radiative transfer equation (RTE). RTE is explicitly written for the reflection function R. The weighting function W is defined as the ratio $\Delta R/2\Delta n$, where $\Delta R = R\left(\lambda,n_0(z)+\Delta n(z_k),\overrightarrow{p_t}\right) - R\left(\lambda,n_0(z)-\Delta n(z_k),\overrightarrow{p_t}\right)$ and Δn is the variation of the ozone density at the level having the altitude z_k. It means that simultaneous determination of R and W requires multiple runs (for all atmospheric heights z_k) of the radiative transfer model (for the profiles $n_0(z)$ and also for $n_0(z)\pm\Delta n(z_k)$). In practice, this procedure can be simplified using the adjoint radiative transfer equation (Rozanov, 2006). Such an approach is followed in this work as well.

We conclude that measured spectra $R\left(\lambda,n_t(z),\overrightarrow{p_t}\right)$ in combination with Eq. (2) allow us to determine $\delta n(z)$ solving the inverse problem. Basically, WF relates the variation of the profile $\delta n(z)$ with the variation of the spectral reflectance as shown in Eq. (2).

We restrict ourselves with the retrieval of the ozone vertical column V defined as follows:

$$V = \int_0^H n(z)dz. \tag{3}$$

where $n(z)$ is an arbitrary ozone vertical profile.

In order to introduce the variation of the vertical column in Eq. (2) we assume that

$$\delta n(z) = Sn_0(z) \tag{4}$$

where S is the constant to be found. Eq. (4) is called the scaling approximation and often used to retrieve the vertical column of absorbing gases in the framework of the differential optical absorption spectroscopy technique. All results given below are valid under assumption that Eq. (14) holds.

Varying Eq. (3) and employing the scaling approximation as given by Eq. (4), we obtain

$$\delta V = \int_0^H \delta n(z)dz = SV_0, \tag{5}$$

where V_0 is the vertical column corresponding to the vertical profile $n_0(z)$ and $\delta V = V_t - V_0$. Substituting further Eq. (4) into Eq. (2), we have

$$R\left(\lambda,n_t(z),\overrightarrow{p_t}\right) = R\left(\lambda,n_0(z),\overrightarrow{p_t}\right) + SW_V(\lambda) \tag{6}$$

where the weighting function W_V for the vertical column is introduced. Namely, it follows:

$$W_V = \int_0^H W\left(\lambda,n_0(z),\overrightarrow{p_t}\right)n_0(z)dz. \tag{7}$$

Thus, using Eq. (6), we find that the difference between the measured and synthetic spectra, which we define as

$$Y(\lambda) = R\left(\lambda,n_t(z),\overrightarrow{p_t}\right) - R\left(\lambda,n_0(z),\overrightarrow{p_t}\right) \tag{8}$$

can be modeled as the product of the known function $W_V(\lambda)$ (see Eq. (7)) and the scaling factor S.

The scaling parameter S and, therefore, δV can be found as the solution of the following minimization problem:

$$\Phi(S) = \left\| Y(\lambda) - SW_V(\lambda) \right\|^2 \rightarrow \min, \tag{9}$$

where $\left\| \quad \right\|^2$ is the norm. The only unknown parameter in Eq. (9) is S. Having retrieved S, the variation of the vertical column can be found according to Eq. (5).

However, the difference $Y(\lambda)$ between two spectra can be caused not only due to the variation of the vertical ozone profile and, therefore, by the variation of the vertical column but also due to the variation of other atmospheric parameters. In our case, to investigate the influence of the uncertainty of the cloud parameters we assume that $\delta n(z)=0$ and, therefore, there is no perturbation of the vertical ozone profile (i.e., $n_t(z)\equiv n_0(z)$). The difference between the measured and synthetic spectra is caused in this case just because biased cloud parameters are taken for the calculation of the synthetic reflectance spectra. In particular, the cloud parameters contained in the vector $\overrightarrow{p_0}$ (which differs from the $\overrightarrow{p_t}$ due to variations of h_{top}, h_{bot} or τ) are used. Thus, Eq. (8) can be rewritten as follows

$$Y(\lambda) = R\left(\lambda, n_t(z), \overrightarrow{p_t}\right) - R\left(\lambda, n_t(z), \overrightarrow{p_0}\right). \tag{10}$$

The change of cloud parameters leads, as a rule, to the change, on one hand, of the reflection function of the Earth–atmosphere system, and, on the other hand, to the change of the photon pathlength distribution (PPD). The first effect explains why the function $Y(\lambda)$ (see Eq. (10)) has a component, which is spectrally neutral or at least smoothly dependent on the wavelength. The change of PPD leads to the change of the absorption bands depths in the ozone spectrum. The spectral signature of this effect is very similar to the spectral signature of the ozone absorption coefficient.

The DOAS (Borell et al., 2004) approach is used to remove the monotonous (without maxima and minima as plotted against the wavelength) component from the total measured signal. As a rule, this component is not known a priori. For this, differential spectra of measured (D_{m}) and synthetic spectra (D_{s}) are introduced as follows:

$$D_m(\lambda) = \ln\left\{R\left(\lambda, n_t(z), \overrightarrow{p_t}\right)\right\} - Q_m(\lambda), \tag{11}$$

$$D_s(\lambda) = \ln\left\{R\left(\lambda, n_0(z), \overrightarrow{p_0}\right)\right\} - Q_s(\lambda). \tag{12}$$

The value of D is sometimes referred to as the differential optical depth (DOD). Here functions $Q_{m(s)}(\lambda)$ can be presented as the polynomials of the third order:

$$Q_m(\lambda) = \sum_{l=0}^{3} a_l^{\mathrm{m}} \lambda^l, Q_s(\lambda) = \sum_{l=0}^{3} a_l^{\mathrm{s}} \lambda^l. \tag{13}$$

Coefficients a_l^{m} can be found as the solution of the following minimization problem:

$$\left\| \ln\left\{R\left(\lambda, n_t(z), \overrightarrow{p_t}\right)\right\} - \sum_{l=0}^{3} a_l^{\mathrm{m}} \lambda^l \right\|^2 \rightarrow \min. \tag{14}$$

The similar equation can be formulated for the determination of a_l^{s} (see Eqs. (12) and (13)).

Let us introduce the difference of DODs:

$$N(\lambda) = D_{\mathrm{m}}(\lambda) - D_{\mathrm{s}}(\lambda). \tag{15}$$

Then the scaling factor S can be found solving the following minimization problem:

$$\overline{\Phi}(S) = \left\| N(\lambda) - S\overline{W}_V(\lambda) \right\|^2 \rightarrow \min. \tag{16}$$

where $\overline{W}_V = W_V(\lambda)R^{-1}\left(\lambda, n_0(z), \overrightarrow{p_0}\right) - Q_W(\lambda)$ and $Q_W(\lambda)$ is the polynomial similar to those given in Eq. (13) but with different coefficients (a_l^W). It follows from Eq. (16):

$$\hat{S} = \frac{\left(N(\lambda), \overline{W}_V(\lambda)\right)}{\left(\overline{W}_V(\lambda), \overline{W}_V(\lambda)\right)} \tag{17}$$

for the value of $\hat{S}$ giving the minimum of $\overline{\Phi}(S)$. Here (,) denotes the scalar product.

Clearly, the minimization problem (16) provides more accurate results for the scaling factor as compared to the problem given by Eq. (9) because unknown but spectrally smooth contribution is removed (see Eq. (11)) and we deal directly with the differential structure in the retrieval procedure. So all results of this work are obtained using Eq. (17). The variation of DOD as given by Eq. (15) is exclusively due to the change in PPD. This is because we introduce no variation in the ozone concentration in this work, i.e., $n_0(z)\equiv n_t(z)$.

Solving the minimization problem as given by Eq. (17), we find the estimation of the scaling parameter, $\hat{S}$, minimizing the given difference between the measured and synthetic differential spectra. In this case the estimated variation of the vertical column as given by

$$\delta\hat{V} = \hat{S}V_0 \tag{18}$$

will be considered as an error of the vertical column determination caused by the usage of the cloud parameters describing by $\overrightarrow{p_0}$ instead of the true vector $\overrightarrow{p_t}$.

In this paper relative errors (in percent) as defined by

$$\varepsilon = \frac{V_t - \hat{V}}{V_t}, \tag{19}$$

where $\hat{V}$ is the retrieved value of the vertical column, are derived and studied. Taking into account that

$$\hat{V} = V_0 + \delta V \equiv V_0 + \hat{S}V_0 \quad (20)$$

and according to our assumption $V_t = V_0$, we have the following expression for the relative error: $\varepsilon = -\hat{S}$, where $\hat{S}$ is given by Eq. (17). This provides a desired relationship between the error of the total ozone due to uncertainty in cloud parameters and the weighting function. The next section is aimed to the application of Eq. (17) for investigations of the total ozone error as retrieved using DOAS technique for unconstrained cloud parameters specified by the vector — parameter $\overrightarrow{p_t}$.

3. Numerical experiments

3.1. Atmospheric model and perturbation scenarios

The following reference atmospheric model is used. The background atmosphere is composed of aerosol and molecules of different gases, which scatter and absorb light. The homogeneous water cloud is placed between heights 5 and 6 km (see Fig. 1). Optical properties of the cloud are calculated using the Cloud C1 model (Deirmendjian, 1969). It is assumed that the cloud optical thickness is 10. Other details of the atmospheric model are given in the Appendix. The surface albedo is assumed to be equal to zero. This model will serve as a reference case and perturbations will be performed with respect to this fixed model. The errors found are characteristic for clouds similar to those specified above. Clearly, Eq. (17) can be used in a similar way to study other types of clouds, including multi-layered cases. However, we restrict our study with this simplest case.

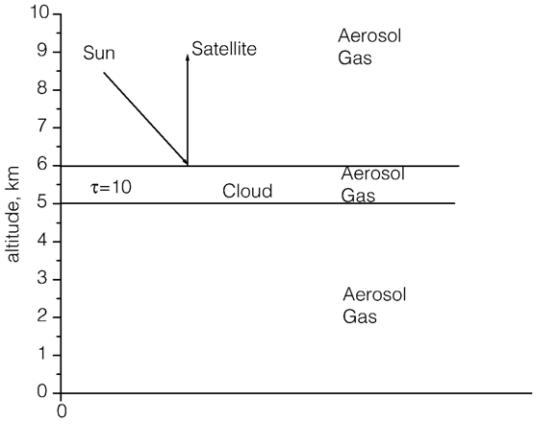

Fig. 1. The assumed cloud position and atmospheric model.

We have performed the following four independent perturbations of the initial atmospheric state with a ground albedo equal to zero:

- The upper cloud boundary is moved upward in steps of 1 km (scenario 1);
- The lower cloud boundary is moved down in steps of 1 km (scenario 2);
- The cloud as a whole (the cloud geometrical thickness Δh is constant) is moved down in steps of 1 km (scenario 3);
- The cloud optical thickness is changed for the fixed cloud position shown in Fig. 1 (scenario 4).

In scenarios 1–3 the cloud optical thickness is kept constant.

3.2. Errors in total ozone due to uncertainties in cloud parameters

The results of calculations of relative errors ε using Eq. (17) in the microwindow 315–340 nm and the SCIATRAN scalar radiative transfer model needed for the calculation of weighting functions (Rozanov et al., 2005) are shown in Fig. 2a for scenarios 1–3 at solar zenith angles 30 and 60° for the nadir observation conditions. It follows that the error ε as introduced above is a monotonous function of the perturbation parameter. For instance, in the case of scenario 1 the error remains negative (too large retrieved total ozone, see Eq. (19)) and increases with the deviation of the cloud upper boundary altitude from its "true" value of 6 km. The error reaches −4% at the solar zenith angle 60°, if there is a bias of +5 km in the cloud top height position. The error is somewhat smaller for smaller solar zenith angles. This underlines the importance of a correct determination of the cloud top height for total ozone retrieval (especially for low sun positions, which is usually the case in polar regions). The bias of +2 km in the cloud top height reduces the error to −1.6% at 60° solar zenith angle. The absolute error is smaller than 1% if the cloud top height is determined with an accuracy of 1 km and $\vartheta_0 = 60°$ (see Fig. 2a). This means that a very precise information on the cloud top altitude (e.g., on an order of meters as provided by lidars) is not required for the determination of total ozone. On the other hand, the use of climatologic values of the cloud top height for a given location may produce quite large errors in the retrieved value of total ozone.

The sign of an error has a quite transparent physical meaning. Indeed, the perturbation of a cloud as specified in scenario 1, hardly changes the shielding

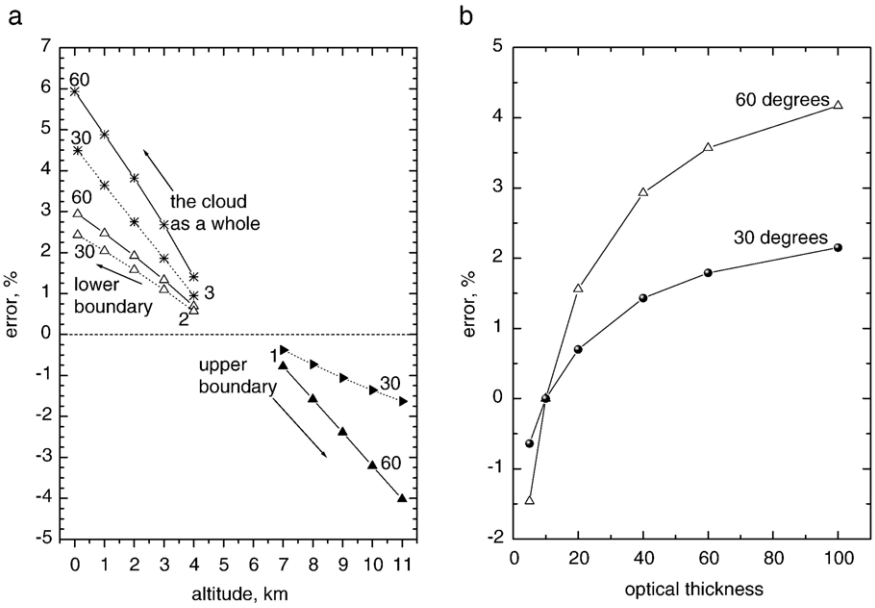

Fig. 2. a. Dependence of the error in the retrieved total ozone on the cloud boundary altitude for scenarios 1, 2, and 3 for the fixed cloud optical thickness 10 and the solar zenith angles 30 and 60°. Curve 1 corresponds to the change in the position of the upper boundary (scenario 1). Curve 2 corresponds to the change in the position of the lower boundary (scenario 2). Curve 3 corresponds to the perturbation of the cloud position without changing its geometrical and optical thickness (scenario 3). b. Dependence of the error in the retrieved total ozone on the cloud optical thickness for the cloud placed between 5 and 6 km for two solar zenith angles. Other details are given in the text.

and albedo effects of a cloud because cloud optical thickness is kept constant. However, a larger portion of atmosphere is incorporated in the cloud then. This means that the depth of absorption bands in the recorded spectrum becomes larger and can be misinterpreted as an enhanced production of ozone $\left(\hat{V}>V_t\right)$. In reality, this is, of course, a solely cloud top altitude uncertainty effect.

There are numerous techniques to determine cloud top heights from a satellite (see a comprehensive review prepared by Rozanov and Kokhanovsky, 2004). This is not the case for the cloud bottom height. However, as it follows from Fig. 2a, the information on the position of the cloud bottom is also of importance for satellite ozonometry especially for a low sun position. We see that the error in the cloud bottom altitude on the order of 2 km leads to an error in the total ozone concentration of 1.3% at $\vartheta_0=60°$ (see curve 2 in Fig. 2a). The error increases to 3% at $\vartheta_0=60°$ if the cloud assumed in the retrieval procedure has a lower boundary at 100 m instead of the "true" value equal to 5 km (for a fixed value of the cloud optical thickness). The influence of the lower boundary position is less important at $\vartheta_0=60°$ for scenario 2 and also for all other scenarios studied (see Fig. 2a,b).

The underestimation of the total ozone for the scenario 3 (see Fig. 2a) is due to the fact that for lower atmospheric layers, the ozone absorption is lower leading to smaller in-cloud absorption effects.

The influence of the uncertainty in the cloud optical thickness and, therefore, cloud albedo on the retrieved ozone vertical column is shown in Fig. 2b (scenario 4) for a fixed cloud position. The "true" cloud has the optical thickness $\tau=10$. The error is zero if one assumes the value of $\tau=10$ in the retrieval procedure. It becomes positive for values of τ in the retrieval algorithm that are larger than the true cloud optical thickness. The opposite is true for smaller τ (see Fig. 2b). These effects are solar angle dependent as shown in Fig. 2b. As in the case of scenarios 1–3, errors are larger for lower sun positions. The underestimation of the total ozone for thicker clouds is due to shielding effect.

Summing up, we conclude that the information on the "true" cloud characteristics like the cloud geometrical thickness, the cloud top height position, and the cloud optical thickness are needed for an accurate estimation of the total ozone from space.

3.3. The validity of Lambertian cloud assumption

In some ozone satellite retrievals procedures, the substitution of a cloud by the impenetrable Lambertian surface is performed (e.g. Klenk et al., 1982, Koelemeijer and Stammes, 1999; Coldewey-Egbers et al.,

2005). Such a technique speeds up the cloud retrieval algorithm used in combination with the ozone retrieval scheme. Then h_{top} is the only cloud parameter to be retrieved from fitting a modeled signal (e.g., O_2 A-band absorption spectra (Rozanov and Kokhanovsky, 2004)) with the measured one. The information on h_{bot} and τ is not available in this case. To study the influence of such a substitution on the accuracy of the total ozone retrieval in a quantitative way, we calculate our synthetic spectra using a Lambertian surface with the spherical albedo 80% instead of the "true" cloud (see, e.g., Koelemeijer and Stammes, 1999). In this case DOD of the synthetic spectra as given by Eq. (12) is replaced by

$$D_s(\lambda) = \ln\left\{R_L(\lambda, n_t(z), h_L)\right\} - Q_L(\lambda), \quad (21)$$

where $R_L(\lambda, n_t(z), h_L)$ is the reflectance at the top of atmosphere with the Lambertian surface positioned at the altitude equal to h_L. The synthetic spectra, $R_L(\lambda, n_t(z), h_L)$ and the corresponding weighting functions have been calculated at the set of $h_L = 3$, 4, 5, and 6 km.

The synthetic spectra corresponding to the "true" atmospheric scenario have been calculated in this case for the set of optical thicknesses of $\tau_t = 5$, 10, 20, 40, 60 and 100. Thus, for each τ_t, errors of the ozone vertical column caused by the substitution of the "real" cloud by the Lambertian surface placed at a given position h_L can be estimated. The results of these calculations are shown in Fig. 3.

It follows from Fig. 3 that, even if the cloud top height is chosen in a correct way (6 km), the error of the retrieved total ozone is in the range [−3%,3%] at $\vartheta_0 = 60°$ depending on the cloud optical thickness of the "true" cloud. We found that the error changes in the interval [−4%,9%] at $\vartheta_0 = 60°$ depending on the position of the Lambertian surface and the cloud optical thickness. Our estimations of the absolute error in the cloud top height retrievals using the Lambertian assumption gives that the cloud top altitude is underestimated by 1–2 km then (Rozanov and Kokhanovsky, 2004). Interestingly, the error is generally lower for thicker clouds, if one accounts for the bias in h_{top} of 1–2 km. Also, the error vanishes for the cloud optical thickness 40, which roughly corresponds to the cloud albedo 80% and the height bias of −1 km. Therefore, the Lambertian surface model can perform quite well in some ozone retrievals schemes. However, the accuracy is uncertain and depends on the cloud characteristics for a given atmospheric state (see Fig. 3). To further reduce the errors due to simplifying assumptions in cloud parameters, an effective spherical albedo for the cloudy scene with a Lambertian cloud top can be retrieved from spectral regions without significant atmospheric absorption, for instance, near 380 nm (e.g. Herman et al., 1997, Coldewey-Egbers et al., 2005) In order to further improve total ozone retrieval more advanced cloud retrieval algorithms have been designed (see, e.g., Rozanov and Kokhanovsky, 2004).

The error range of the total ozone retrieval caused by the different position of the Lambertian surface can be smaller if the position of the Lambertian surface, h_l^*, is found by fitting corresponding O_2 A-band synthetic spectra with respect to the measured spectra corresponding to the "true" cloud. We show h_l^* retrieved for different τ_t by asterisks in Fig. 3. We found that the Lambertian surface assumption gives smaller values of h_{top} as compared to the "true" cloud top height. The bias ranges from 0.47 to 1.17 km depending on τ and ϑ_0 (see Fig. 3). This is in a good agreement with the previous estimations (Rozanov and Kokhanovsky, 2004).

It follows from Fig. 3 that the error of total ozone retrieval using the derived position of Lambertian surface from O_2 A-band reflectance is in the range [−2.5%,4.5%] at the solar zenith angle of 60° and it is in the range [−0.5%,3%] at $\vartheta_0 = 30°$. As expected, maximum errors in the total ozone retrieval are found in the case of small optical thickness of the "true" cloud.

Fig. 3 confirms our earlier findings (see Fig. 2) that the errors are generally smaller for smaller solar zeniths angles. It must be underlined that values of the solar zenith angle ϑ_0 are usually quite low both in Arctic and Antarctic, where the ozone hole is located during spring. The cloud characteristics in these polar regions are of special value as compared to tropics, where ϑ_0 is small. We also found that errors for overhead sun ($\vartheta_0 = 0°$) slightly differ from those for the sun at $\vartheta_0 = 30°$.

4. Summary and discussions

We have studied the influence of uncertainty in the parameters of the idealized horizontally and vertically homogeneous clouds on total ozone retrievals from space using DOAS. It is confirmed that the cloud optical thickness, the cloud top height, and the cloud geometrical thickness must be known in advance for an accurate ozone concentration retrievals performed by optical instruments on satellite platforms. The Lambertian cloud reflection assumption sometimes used in ozone satellite retrievals may bias results significantly depending on the type of cloudiness. Incorrect information on cloud characteristics can cause errors of up to 6% as shown in Fig. 2.

We found that the error is positive for clouds with underestimated cloud top positions as compared to the "true" cloud top altitude h_{top}. The error depends on the

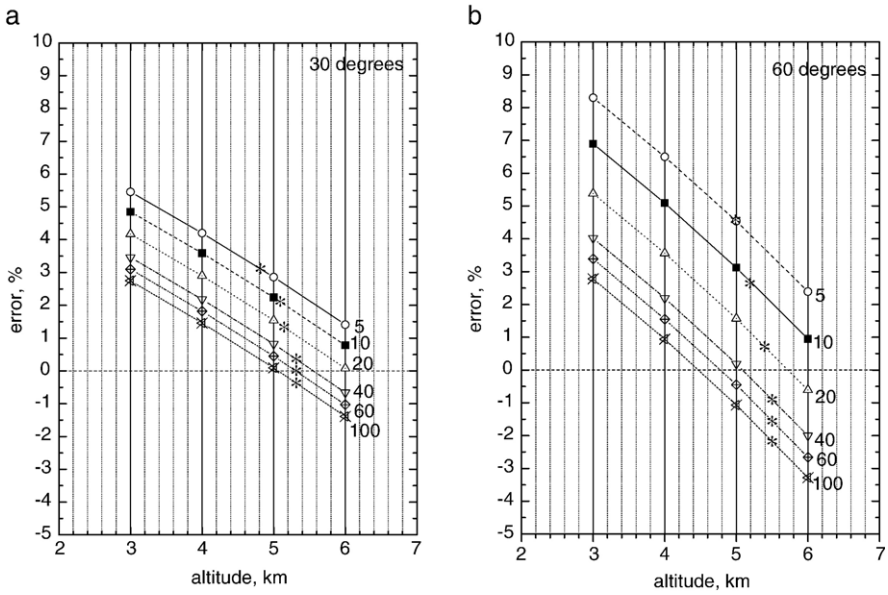

Fig. 3. a. Dependence of the error in the retrieved total ozone on the altitude of the Lambertian surface (with the albedo 80%) position at different heights at 30° solar zenith angle. The "true" cloud top height is located at 6 km. The "true" cloud optical thickness is varied in the range 5–100. The asterisks give the positions of the retrieved Lambertian surface using simulated O_2 A-band measurements. b. The same as in Fig. 3a except for the solar zenith angle 60°.

solar zenith angle and it is below 6% for all cases studied in this work. Taking into account that total ozone corresponding to the used vertical ozone profile is equal to 302 DU, we conclude that the underestimation of total ozone is around 18 DU at maximum, if h_{top} is underestimated. In particular, we found that the overestimation of the cloud top height by 5 km, which is possible, if climatologic cloud top positions are used, gives an error of −4% or overestimates total ozone by approximately +12 DU for the case studied.

The error ε is linear with respect to h_{top}. It means that Fig. 2a can be also used to estimate errors in cases, when the "true" cloud is positioned at yet another atmospheric level (see, e.g., Liu, 2002) and not in the range of 5–6 km as assumed in this study. This enables the extension of our results for a broader class of cloudy media.

The combination of retrieving Lambertian reflecting boundary height and effective albedo (Coldewey-Egbers et al., 2005) as part of the satellite ozone retrieval may reduce the bias to first order, however, there is a strong need for implementing more advanced cloud retrieval algorithms in ozone retrievals codes. This becomes even more important for methods that derive tropospheric ozone from total column measurements using cloud information (Borrell et al., 2004).

The general theoretical technique as presented here can be used to investigate the impact of errors due to clouds (and also other atmospheric state parameters) on the accuracy of other trace gas vertical columns and profiles, including NO_2, SO_2, CH_4, and other species in the framework of the scaling approximation given by Eq. (4).

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Appendix A. Atmospheric model

The following atmospheric model has been used in this study. The atmosphere was divided into 4 layers positioned between 0 and 2 km, 2 and 10 km, 10 and 30 km, and 30 and 60 km with different aerosol properties attributed to each layer as specified by Kneizys et al. (1996). This allows to account at least in the first approximation for the aerosol stratification (e.g., boundary layer aerosol, tropospheric aerosol, and stratospheric aerosol). The most important influence on the simulated signal comes from the lower aerosol layer. This is due to the fact that aerosol sources are mostly located at the surface in the lower atmosphere (e.g., water vapour and other trace gases). The total aerosol optical

thickness τ is assumed to be equal to 0.2. We are mostly concentrated on the influences of clouds on the retrieval of the total ozone vertical density in this paper. Therefore, the aerosol phase function $p_a(\theta)$ was given by the Heney–Greenstein approximation:

$$p_a(\theta) = \frac{1-g^2}{(1+g^2-2g\cos\theta)^{3/2}},$$

where θ is the scattering angle and g is assumed to be equal to 0.6 in the retrieval procedures described in this work. The phase function for the molecular scattering was taken in the form:

$$p(\theta) = 0.75\left(1+\cos^2\theta\right).$$

It was assumed that water clouds are positioned at various levels in the terrestrial atmosphere. The cloud optical thickness has been varied. However the cloud phase function was fixed. It was calculated using Mie theory as specified by Kokhanovsky and Rozanov (2004) for the gamma droplet size distribution (DSD) with the effective radius 6 μm and the coefficient of variance of droplet size distribution equal to 37%. The absorption of light by cloud droplets is neglected.

We have used the vertical profiles of temperature, pressure, and ozone concentration for the northern hemisphere at latitude 45N, June 15th incorporated in SCIATRAN (Max Planck Institute (Hamburg) models). The total ozone concentration was assumed equal to 302 DU. The probability of photon absorption $\beta = 1-\omega_0$ for the model used is shown in Fig. A1 as the function of the altitude. Here the single scattering albedo ω_0 is the ratio of the atmospheric scattering and atmospheric extinction coefficients.

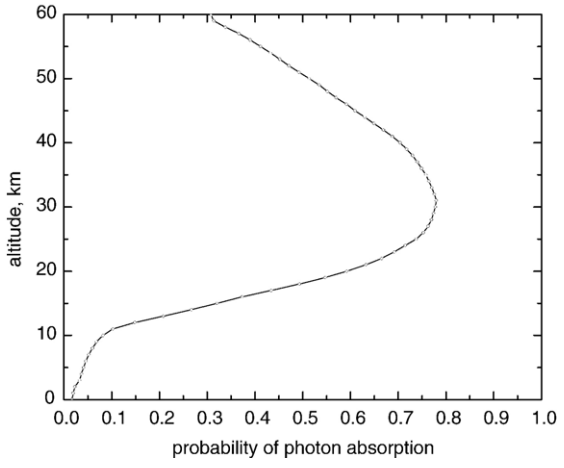

Fig. A1. The vertical profile of the probability of photon absorption (or co-scattering albedo $1-\omega_0$) at 320 nm for the cloudless atmosphere.

Further details of the model used are given by Kokhanovsky and Rozanov (2004). SCIATRAN scalar radiative transfer code (Rozanov et al., 2005) used in this paper for the calculation of spectral TOA radiances and weighting functions is freely available at www.iup.physik.uni-bremen.de/sciatran.

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